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We report the first account of a cluster fragmentation study involving high energy cluster-cluster collisions in which all the fragments of each collision occurring in the experiment are mass analysed on an event by event basis. This allows an unbiased look at the true nature of fragmentation including a statistical analysis in terms of fluctuations in the fragment size distribution.

Fragmentation of finite size systems is a wide spread phenomenon in nature, including such diverse phenomena as the break-up of submicroscopic objects¹ or collisions between asteroids². Despite intensive research in various fields of science and technology complete analysis and understanding of fragmentation has not yet been achieved. Nevertheless, there is the recognition that general features of this phenomenon are rather independent of the actual system and its underlying interaction forces³. Thus it is highly desirable to be able to study a model system in as much detail as possible in order to arrive at sound conclusion which may be applied (and compared) to a larger range of objects. So far most of the studies concerning fragmentation have been able to determine, besides other properties, average fragment size distributions without information on the size distribution of a single fragmentation event. In contrast, here we report the first account of a cluster fragmentation study involving collisions of high energy hydrogen cluster ions with fullerenes in which all the fragments of all collisions occurring in the experiment are mass analysed on an event by event basis using a novel multi-coincidence technique⁴. This allows an unbiased look at the true nature of fragmentation including a statistical analysis of the data set consisting of 6000 events in terms of fluctuations in the fragment size distribution.

The experimental characterization of the cluster fragmentation, not only by the average fragment size distribution but also by a statistical analysis of the fragmentation events, has become possible owing to a recently developed multi-coincidence technique for the detection of ionized and neutral fragments⁴. In short, mass selected hydrogen cluster ions with an energy of 60 keV/amu are prepared in a high-energy cluster ion beam facility consisting of a cryogenic cluster jet expansion source combined with a high performance electron ionizer and a two-step ion accelerator. After momentum analysis by a magnetic sector field, the mass selected and pulsed high energy projectile beam (pulse length of 100 ns; repetition frequency of 1 Hz) consisting in the present study of H_{25}^+ cluster ions, is collimated by two apertures ensuring an angular dispersion of about ± 0.8 mrad. This cluster ion projectile beam is crossed perpendicular by a C_{60} effusive target beam (see Fig.1) produced by evaporation of pure C_{60} powder in a single-chamber molybdenum oven at about 950 K. One meter behind this collision region the high energy hydrogen collision products (neutrals and ions) are passing through a magnetic sector field analyser approximately 0.3 μ s after the collision event. Both, undissociated primary H_{25}^+ cluster projectile ions and neutral and charged fragments resulting from reactive collisions are then detected with a multi-detector device consisting of an array of surface-barrier detectors located at different positions at the exit of the magnetic analyser (see Fig.1). This allows us to record simultaneously neutral and charged fragments detected in coincidence for each single collision event (for more experimental details see Ref. 4 and references therein) irrespective of the nature of the collisional interaction, i.e., small or large impact parameters leading to rather "gentle" or "violent" collisions. Because of the short time of the collisional interaction the internal motion of the cluster constituents during the collision is negligibly small, therefore in each case the excitation can be thought as being due to a sudden

perturbation followed by the cluster fragmentation governed by the energy deposited.

Previous studies including also hydrogen cluster ion-helium atom collisions showed analogous to the case of nuclear collisions two different types of averaged fragment ion mass distributions. For the helium target where the energy deposited in the hydrogen cluster ion during the collision is on average smaller a bimodal distribution with increasing probability for the production of smaller and larger fragments has been observed⁵, whereas for the fullerene target where the energy deposited is on average larger we observed a single power-law distribution increasing with decreasing fragment size⁶. Moreover, from first studies concerning the multiplicity of fragments⁶ it is clear that on average small fragments are due to the violent collisions involving a large amount of energy transfer and large fragments are due to the gentle type of interactions. Nevertheless, in both target cases the size distribution of the smaller fragments can be described by a power-law fall-off independent of the cluster size and similar to what is known for nuclear collisions⁷⁻¹⁰.

Many phenomena in nature^{11,12} and human life^{13,14} are governed by power laws. Moreover, there exists a strong similarity between the fragment size distribution and the predictions of certain models describing critical phenomena. The most famous is the Fisher droplet model¹⁵ which allows to calculate the droplet size distribution in a vapor. At the critical temperature the resulting distribution $f(p)$ in sizes p is proportional to $p^{-\tau}$ and the predicted exponent of 2.23 is close to the values observed for nuclear⁷⁻¹⁰ and cluster fragmentations^{5-6, 16-18}. Although these observations have been taken as a strong hint for the occurrence of critical behaviour in a finite system reminiscent of a second order phase transition in an infinite system. it cannot be considered as definitive proof. Bond percolation models have also been used to describe nuclear fragmentation and to simulate critical behaviour predicting, besides the existence of a power law, additional properties of the fragment size distribution. These secondary characteristics (including Campi plots, multi-factorial moment analysis, etc.¹⁹⁻²¹) are, however, only accessible via event by event data analysis as performed in the present experiment.

Thus, as an example following Campi¹⁹ we plot and analyse the present results in the following way: In Fig.2 upper part we plot the average size P_{\max} of the largest fragment produced in a single event (normalized to the number of constituents, 25, in the primary hydrogen cluster ion projectile) versus the multiplicity m (normalized as done for P_{\max} to the size of the system). In comparison we show the corresponding results obtained from a three dimensional bond percolation model with 125 sites¹⁹. As the fluctuations in fragment size distributions are largest near the critical point it is interesting to also plot as a measure for these fluctuations the standard deviation of P_{\max} versus the normalized multiplicity (lower part of Fig.2). The following observations and points with respect to these results are noteworthy:

- (1) First of all there is a remarkable agreement in the overall shape of the functions

obtained in the cluster collision experiment on the one hand, and from the percolation model on the other hand. The shift of the resonance-like peak in the standard deviation function is due to the size difference of the two systems.

(2) We have to point out that for a one-dimensional percolation model different results are obtained¹⁹ with no resonance-like peak in the standard deviation function and therefore with no indication of a finite-size phase transition.

(3) An experimental data set available for such an analysis for nuclear fragmentation, i.e., a nearly complete fragment charge analysis of 1 GeV/amu Au ions bombarding an emulsion²² of about 400 events²⁰, exhibits a similar behaviour¹⁹ as the present cluster collision data and the three-dimensional percolation model. It should be mentioned, however, that according to DeAngelis et al.²⁰ this nuclear fragmentation data set is very likely biased towards low multiplicity events as the experimental set-up records only those events in which all 79 charges are detected (see also another recent multifragmentation experiment with gold nuclei where the detector system permitted exclusive event reconstruction of nearly all charged reaction products²³). In contrast the present cluster collision experiment (with about 6000 events) includes all events and the comparison with the percolation model is unbiased from the detection probability.

Several conclusions (which are also supported by additional use of the other momentum analyses not discussed here) may be drawn from the presented results.

The close resemblance between the results from percolation and the present cluster collision results constitutes a further step in ascertaining the occurrence of critical behaviour in these finite size collision systems: percolation is thought to provide a useful method to study phase transitions due to the similarities in the behaviour with systems exhibiting second order phase transitions, such as liquid-vapor systems at the critical temperature²⁴. A particular advantage of this comparison is the fact that according to Campi²⁵ it is possible to compare in this way nuclear collision (and also cluster collision) data with results of percolation on finite size lattices of a similar size and thus to avoid the problem of the finite size of these systems.

Finally, quite different objects such as nuclei and molecular clusters show similar fragmentation behaviour not only in the inclusive fragment size distribution but also in the event by event type analysis. This is in accordance with a recent scaling prediction concerning large fluctuations in finite systems^{21,26}. The application of the predicted characterizations of fragment size distributions²⁷ is demonstrated here for the first time for cluster fragmentation data (see also a theoretical study by Lutz and coworkers²⁸). However, besides the usefulness of this approach for microscopic systems like nuclei (fermi scale) or clusters (nanometer scale) it might be also applicable on a more universal scale including macroscopic phenomena.

Having obtained the present far reaching conclusions based on a statistical treatment, the question arises whether another approach such as the reductionism one - i.e., the dynamics of complex systems by considering out its most fundamental constituents and their interactions - could lead to additional insights in a similar way as, at the end of the last century, the success of the thermodynamic approach did not exclude the discovery of the microscopic laws. So, the

success of the statistical approach presented here may serve as stimulus to improve the understanding of the dynamics in complex systems when the correlation length between the constituents is of the same order of magnitude than the size of these systems.

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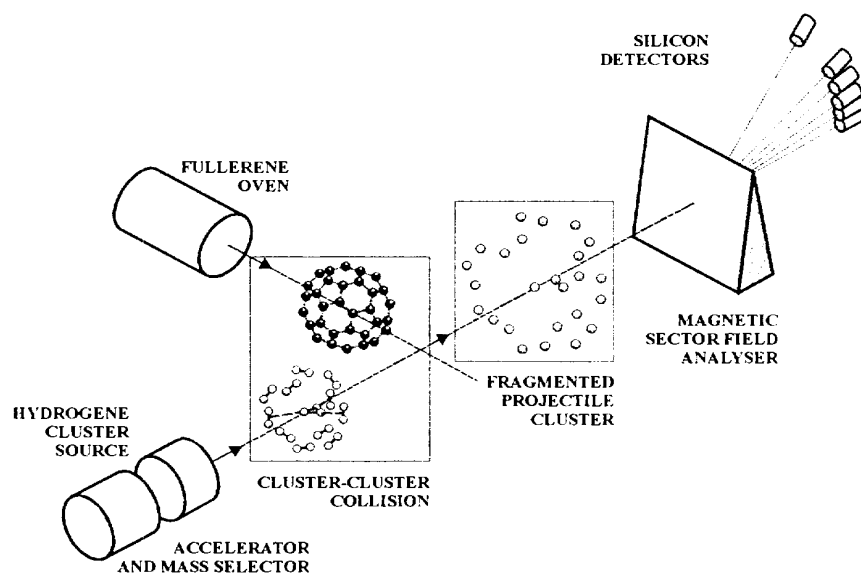
Figure Captions.

Fig.1 Schematic view of the experimental set-up. Mass selected hydrogen cluster ions of 60 keV/amu energy are crossed perpendicularly by an effusive C_{60} beam in the collision region. Neutral and charged hydrogen fragments are passing through a magnetic sector field analyser one meter behind this collision region and then are detected in coincidence with a multi-detector device consisting of a number of surface-barrier detectors located at different positions at the exit of the analyser. For each fragmented cluster, the size and the number of the different fragments are recorded. Thus, for each fragmentation event, the size of the largest fragment and the total number of fragments which is called multiplicity are available for the statistical analysis as presented in Fig. 2.

Fig.2 Comparison of the fragmentation behaviour for two different systems: hydrogen clusters versus 3D bond percolation lattice. In both cases, from the break-up in two fragments (low multiplicity) to the complete desintegration (high multiplicity), the fragmentation phenomenon exhibits a transition with an increase of the fluctuations.

Upper part a): Average size P_{\max} of the largest fragment produced in a single event (normalized to the projectile size 25 or to the number of sites 125 of the cubic lattice, respectively) versus the normalized multiplicity m for the three-dimensional percolation model (open circles) and the present experimental cluster fragmentation results (full circles).

Lower part b): Normalized standard deviation (fluctuation) of P_{\max} given in the upper part versus m . The normalized standard deviation is defined as $\sqrt{\langle P_{\max}^2 \rangle - \langle P_{\max} \rangle^2}$.



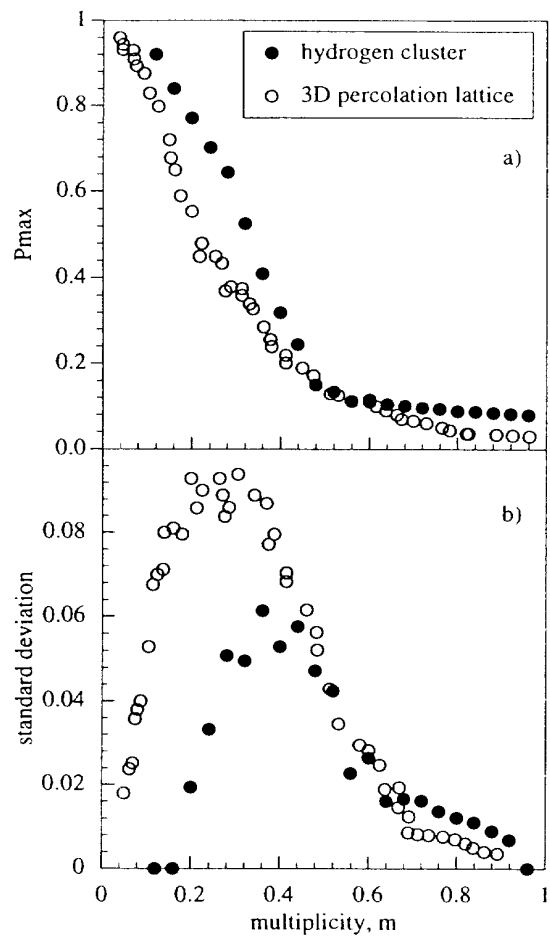


Figure 2: B. Farizon et al.